

## LA-UR-19-20721

Approved for public release; distribution is unlimited.

Title:                   Gamma-ray Analysis of the Pressurized-Water-Reactor and  
Boiling-Water-Reactor Assemblies

Author(s):           Vo, Duc Ta

Intended for:       Workshop Presentation

Issued:               2019-01-30

---

**Disclaimer:**

Los Alamos National Laboratory, an affirmative action/equal opportunity employer, is operated by Triad National Security, LLC for the National Nuclear Security Administration of U.S. Department of Energy under contract 89233218CNA000001. By approving this article, the publisher recognizes that the U.S. Government retains nonexclusive, royalty-free license to publish or reproduce the published form of this contribution, or to allow others to do so, for U.S. Government purposes. Los Alamos National Laboratory requests that the publisher identify this article as work performed under the auspices of the U.S. Department of Energy. Los Alamos National Laboratory strongly supports academic freedom and a researcher's right to publish; as an institution, however, the Laboratory does not endorse the viewpoint of a publication or guarantee its technical correctness.

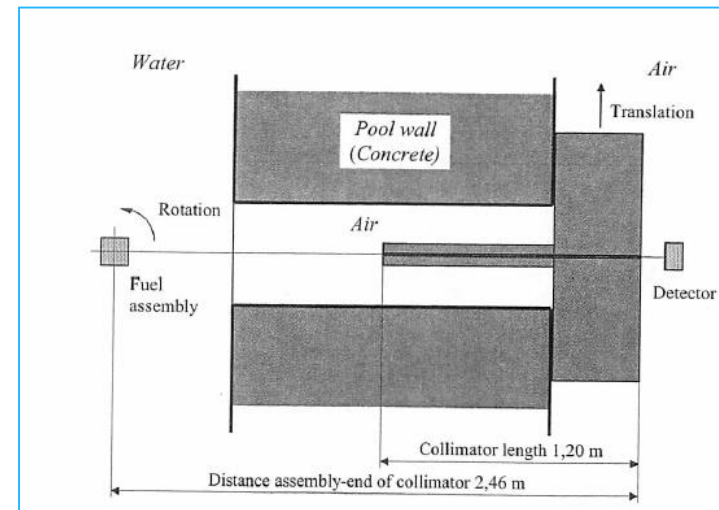
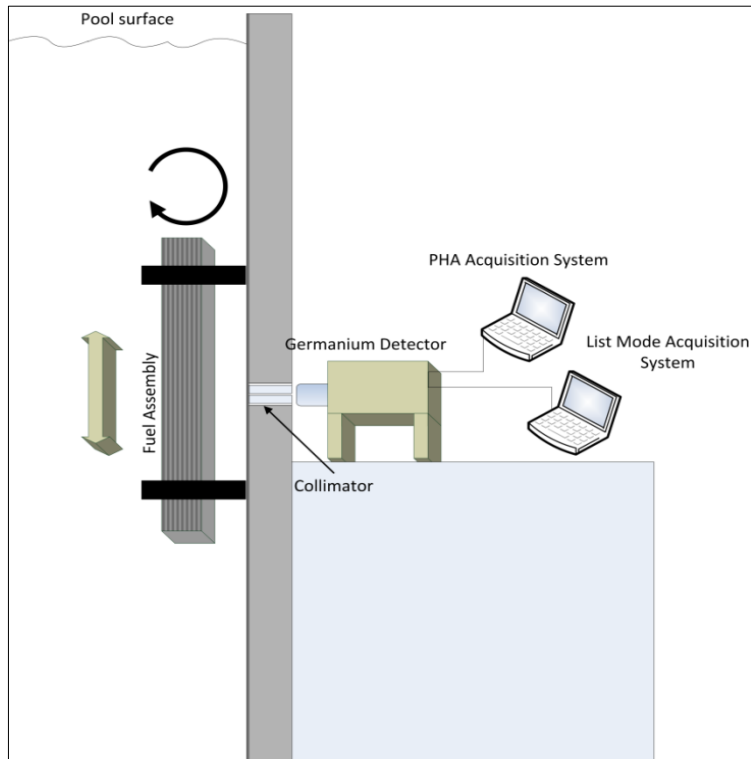
# **Gamma-ray Analysis of the Pressurized-Water-Reactor and Boiling-Water-Reactor Assemblies**

Duc Vo

Los Alamos National Laboratory

# Measurement setup

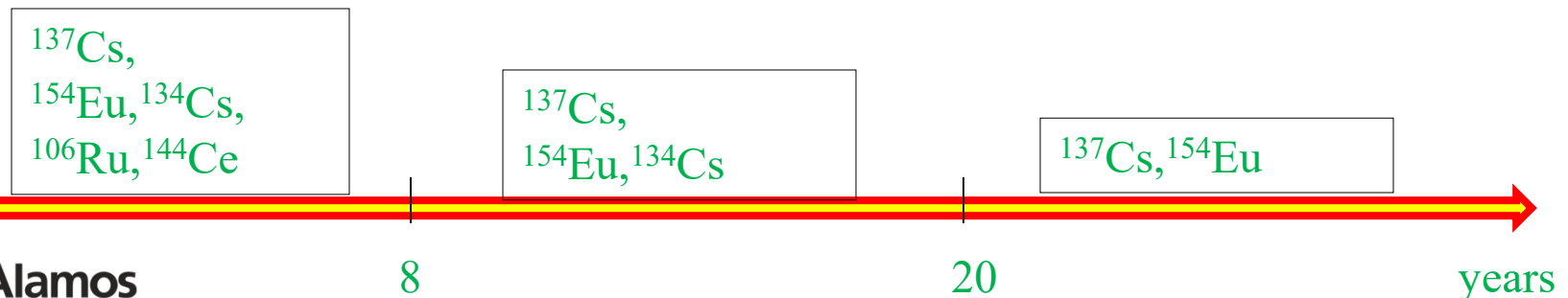
- All measurements were done at Sweden's Central Interim Storage Facility for Spent Nuclear Fuel (Clab)



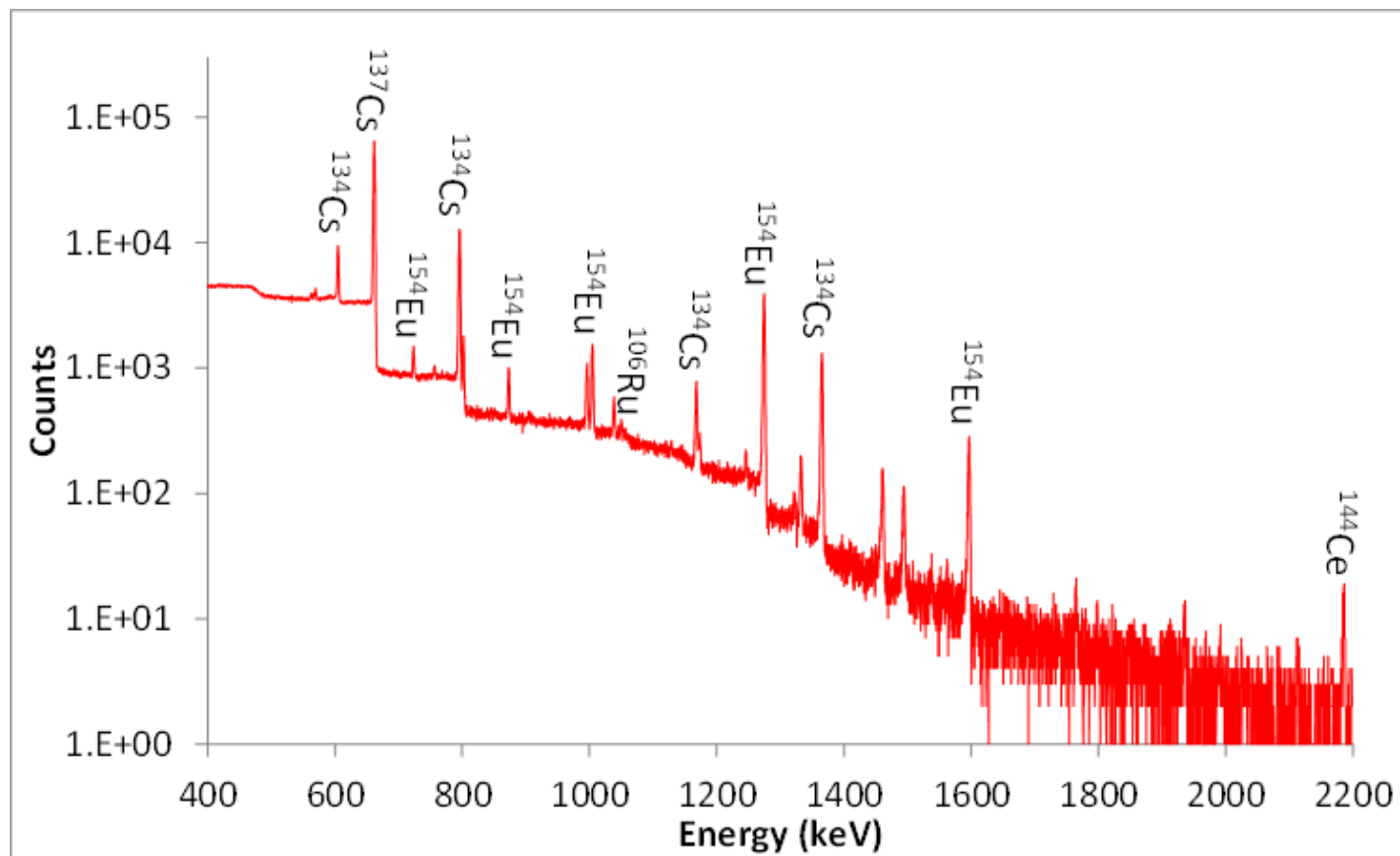
# Main isotopes from nuclear spent fuel

Fission Product Isotopes	Half-life (years)	Cooling time limit (years)
$^{137}\text{Cs}$	30.1	
$^{154}\text{Eu}$	8.5	<100
$^{134}\text{Cs}$	2.1	<20
$^{106}\text{Ru}$	1.0	<8
$^{144}\text{Ce}$	0.8	<8

- $^{137}\text{Cs}$ , and mass isotopic ratios to  $^{137}\text{Cs}$  are used.
- Model functions will be used to extract IE, BU, CT.



# A sample spectrum (CT = 7.5 y)



# FRAM code: extracting isotopic ratios

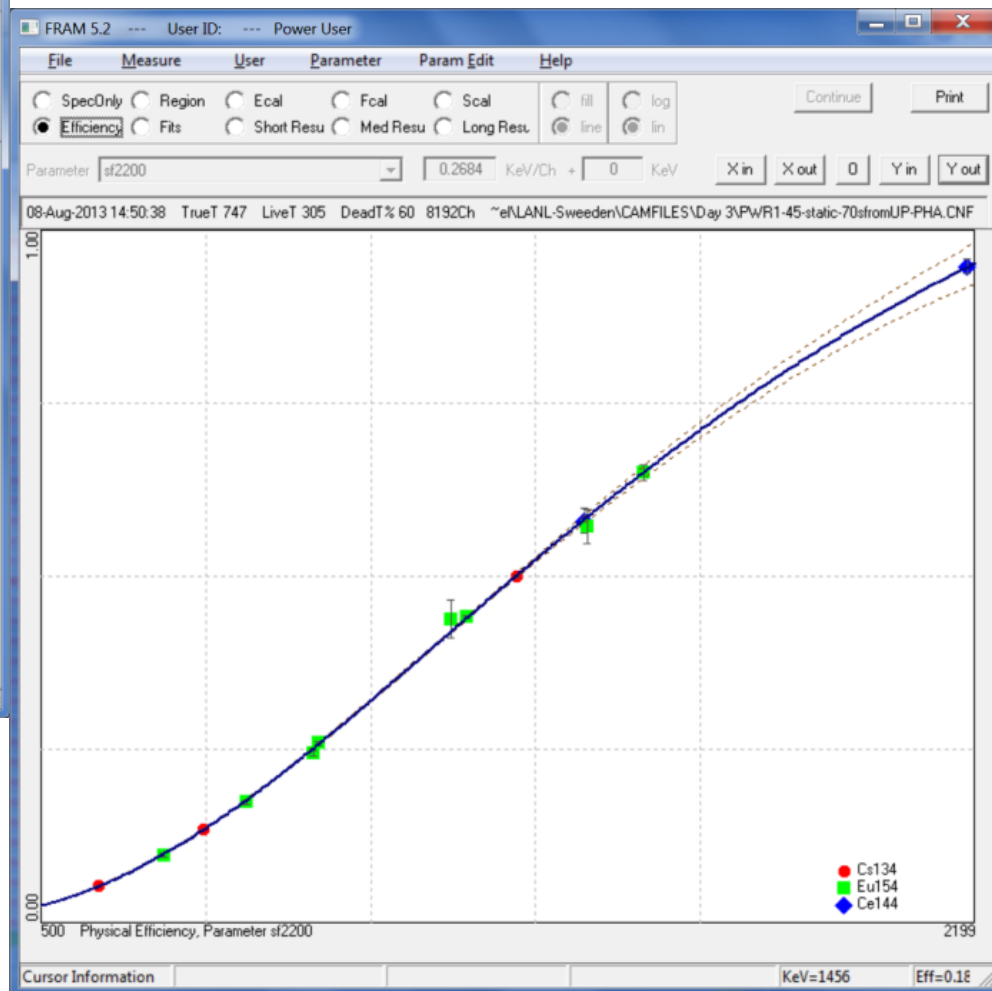
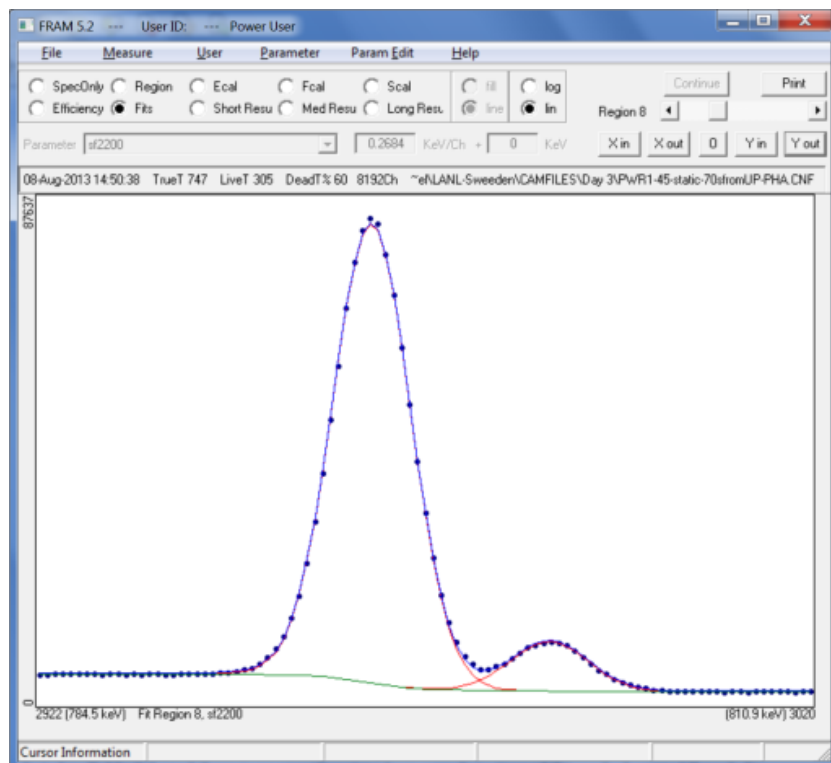
**FRAM** (Fixed energy Response function Analysis with Multiple efficiencies):

- Code designed primarily for plutonium and uranium isotopic analysis.
- Self-calibrates using several peaks and does not need external calibration.

FRAM can also be used to calculate the *ratio of one isotope to that of another isotope*. These can be any isotopes, not necessarily those of plutonium or uranium.

- FRAM works by fitting various regions of the spectrum to extract peak areas; and from those the total efficiency and the relative efficiency.
- Atom ratio of isotope *i* to isotope *k*:
$$\frac{N^i}{N^k} = \frac{C(E_j^i)}{C(E_l^k)} \times \frac{T_{1/2}^i}{T_{1/2}^k} \times \frac{Br_l^k}{Br_j^i} \times \frac{Re(E_l)}{Re(E_j)}$$
- The use of an efficiency ratio removes the need of **reproducible set up** to determine the isotopic ratios

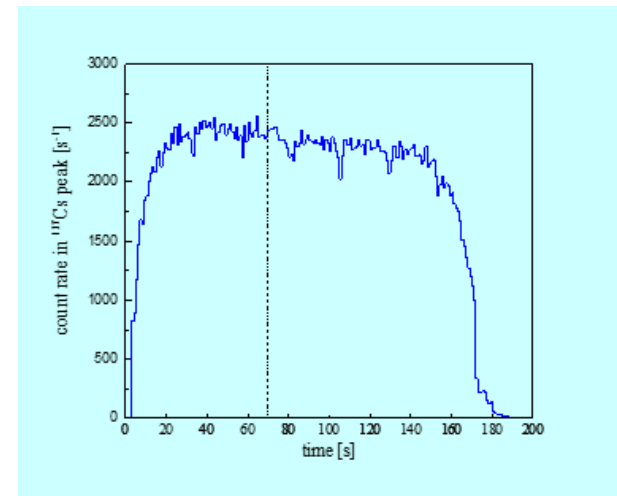
# Peak fitting and Relative efficiency





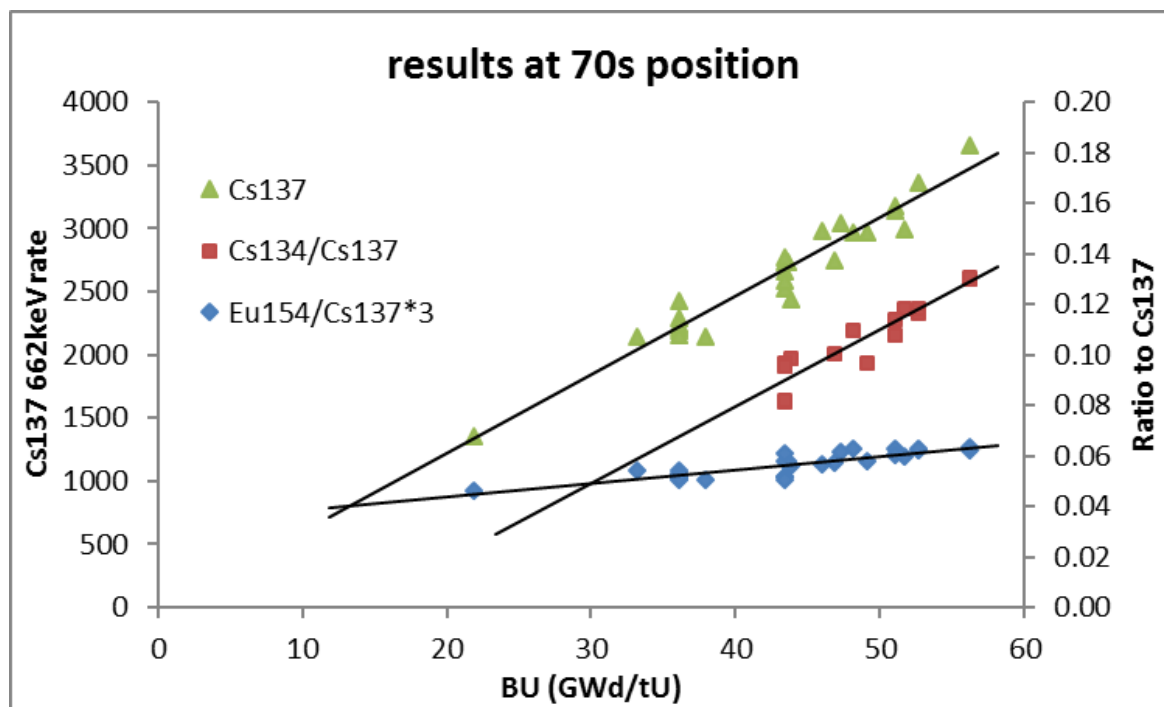
# PWR assemblies measurement campaigns

- **August 2013:**
  - 25 assemblies were measured at 45°, and position 70s (~150 cm from bottom. Assembly length ~ 366 cm).
  - PWR9 spectra acquired along its length from 30s to 165s.
  - PWR16 were measured from 4 corners
  - PWR20 were measured from 3 corners
- **October 2014:**
  - Different detector system and filters.
  - The same 25 assemblies were measured.
  - All 4 corners were measured at position 120 cm from top of assembly.



# FRAM fit results (August 2013)

Distributions of the 662 keV peak rates,  $^{154}\text{Eu}/^{137}\text{Cs}$ , and  $^{134}\text{Cs}/^{137}\text{Cs}$  as functions of the BU for the measurements the continuous or nearly continuous burned assemblies. The data have been adjusted to **CT = 0**.



Data suggest correlation as product of the CT in exponential form and BU as linear or power law function

# Analysis

---

- We fitted the  $^{137}\text{Cs}$ ,  $^{154}\text{Eu}/^{137}\text{Cs}$ ,  $^{134}\text{Cs}/^{137}\text{Cs}$ ,  $^{106}\text{Ru}/^{137}\text{Cs}$ , and  $^{144}\text{Ce}/^{137}\text{Cs}$  information of the spectra to the equations

$$^{137}\text{Cs} = (aBU^b)e^{-\lambda_{137}CT}$$

$$\frac{^{154}\text{Eu}}{^{137}\text{Cs}} = (cBU + d)e^{-(\lambda_{154} - \lambda_{137})CT}$$

$$\frac{^{134}\text{Cs}}{^{137}\text{Cs}} = (eBU + f)e^{-(\lambda_{134} - \lambda_{137})CT}$$

$$\frac{^{106}\text{Ru}}{^{137}\text{Cs}} = (gBU + h)e^{-(\lambda_{106} - \lambda_{137})CT}$$

$$\frac{^{144}\text{Ce}}{^{137}\text{Cs}} = (iBU + j)e^{-(\lambda_{144} - \lambda_{137})CT}$$

# BU and CT analysis results

---

- From the fits we obtained the coefficients  $a - j$ .
- With these coefficients known, the **BU** and **CT** can be determined from the  $^{137}\text{Cs}$  and  $^{137}\text{Cs}/^{137}\text{Cs}$  ratio information extracted from a measurement.
- With 2 unknowns **BU** and **CT** to be determined, at least 2 known parameters ( $^{137}\text{Cs}$  and  $^{154}\text{Eu}/^{137}\text{Cs}$ , or  $^{154}\text{Eu}/^{137}\text{Cs}$  and  $^{134}\text{Cs}/^{137}\text{Cs}$ ) are needed.
- The equations are difficult to solve analytically. So we fit the data to obtain **BU** and **CT**.

# IE analysis results

---

- The **IE** is determined from the **BU**.
  - *The need of power plant to maximize profit can be seen as the need to maximize BU for a given IE (correlation expected)*
  - *Using the data provided from the fuel/reactor owner, we found a simple relationship for all measured assemblies*

$$IE = 0.31 \cdot BU^{0.67}$$

- *The inverse function  $BU = 6 \cdot IE^{1.5}$  looks like an “unpublished” guidance that set the maximum BU to be reached with the fuel per each IE.*

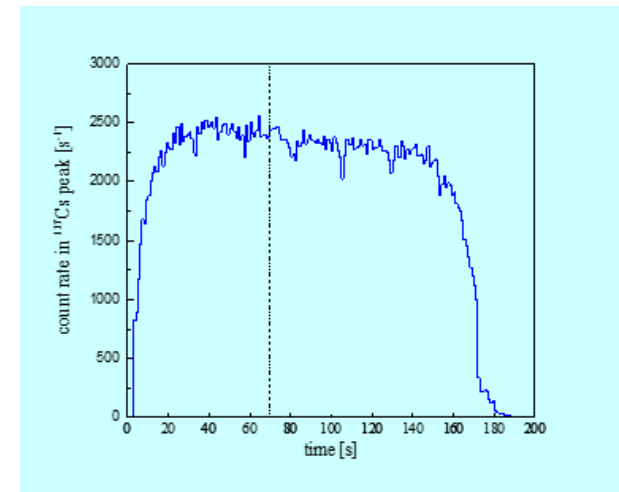
# Analysis results: measurements along the length of the assembly (PWR9)

	BU	CT	IE
Declared	45.8	5.97	3.71
30s mark	45.0	6.38	3.70
70s mark	45.3	6.38	3.72
90s mark	45.6	6.38	3.73
120s mark	46.1	6.38	3.76
150s mark	44.7	6.38	3.69
155s mark	44.0	6.38	3.65
160s mark	39.1	6.38	3.38
165s mark	28.5	6.38	2.75

Remark: all the measurement point across the length has THE SAME CT

The 30s – 120s spectra were analyzed using  $^{137}\text{Cs}$ ,  $^{154}\text{Eu}/^{137}\text{Cs}$ , and  $^{134}\text{Cs}/^{137}\text{Cs}$  information.

The 150s – 165s spectra analysis did not use  $^{137}\text{Cs}$  information (because we were on the edge and the efficiency is not correct).



# Analysis results:

## 4 corners of an assembly (PWR16)

---

	BU	CT	IE
<b>Declared</b>	<b>40.4</b>	<b>17.15</b>	<b>3.60</b>
45°	41.0	17.02	3.48
135°	43.3	18.79	3.61
225°	40.8	18.38	3.48
315°	37.1	15.52	3.27
<b>Average</b>	<b>40.6</b>	<b>17.43</b>	<b>3.46</b>
%Bias (ave/dec-1)	<b>0.36%</b>	<b>1.63%</b>	<b>-3.91%</b>

The spectra were analyzed using  $^{137}\text{Cs}$ ,  $^{154}\text{Eu}/^{137}\text{Cs}$ , and  $^{134}\text{Cs}/^{137}\text{Cs}$  information.

# Analysis results: 3 corners case (PWR20)

	BU	CT	IE
<b>Declared</b>	<b>34.0</b>	<b>27.19</b>	<b>3.10</b>
45°	33.1	27.57	3.03
45° overnight	33.1	28.09	3.03
225°	36.8	27.98	3.25
315°	33.8	27.06	3.07
<b>Average</b>	<b>34.2</b>	<b>27.67</b>	<b>3.10</b>
%Bias (ave/dec-1)	<b>0.47%</b>	<b>1.78%</b>	<b>-0.14%</b>

The spectra were analyzed using  $^{137}\text{Cs}$  &  $^{154}\text{Eu}/^{137}\text{Cs}$  information.



# Remarks (3 & 4 corners)

---

- BU's of the 4 corners of an assembly are slightly different due to position/facing during the burning in the reactor, and thus the average results present better accuracy.
- The assembly may be not perfect straight, and this affects the  $^{137}\text{Cs}$  intensity. (1 cm water reduces the transmission of the 662 keV gamma ray by 8%.) Averaging the results minimizes this potential bias.

# October 2014 campaign

---

- The same 25 PWR assemblies were re-measured
- The setup was different:
  - Detector
  - Electronics
  - Filter
  - Measurement position
- Use the same equations and coefficients of the 2013 measurements except the coefficient *a* for  $^{137}\text{Cs}$  intensity was adjusted due to different setup

# October 2014 campaign: Results

Assembly	BU	CT	IE	Assembly	BU	CT	IE
PWR1	0.0%	-0.7%	0.1%	PWR13	-5.2%	-1.4%	5.0%
PWR2	2.7%	2.8%	2.1%	PWR14	-1.9%	-3.1%	-2.4%
PWR3	-0.2%	-1.4%	4.8%	PWR15	-2.6%	-7.1%	21.3%
PWR4	-11.0%	-8.4%	-10.3%	PWR16	0.9%	-0.3%	-3.5%
PWR5	-14.9%	-11.6%	-13.1%	PWR19	-2.0%	3.5%	-3.0%
PWR7	-7.3%	-5.9%	-11.2%	PWR20	0.8%	-1.8%	0.1%
PWR8	-0.9%	-2.4%	10.5%	PWR21	-0.7%	-1.0%	-0.9%
PWR9	0.2%	1.4%	1.2%	PWR22	0.2%	0.6%	4.2%
PWR10	1.9%	0.5%	-1.0%	PWR25	-1.9%	-0.2%	1.3%
PWR12	11.8%	8.6%	17.1%				

Deviations (measured/declared -1) of the BU, CT, and IE

- PWR4 and PWR5: a spacer was in view of the detector in the 2014 measurements
- PWR12: CT ~ 26 y,  $^{154}\text{Eu}/^{137}\text{Cs}$  error ~ 7%

# BWR assemblies measurement campaigns

---

- **March 2014:**
  - 17 BWR assemblies were measured
  - For each assembly, a corner (45°) was measured.
  - All were measured at positions 92 cm , 187 cm, and 281 cm (assembly length ~370 cm).
  - 3 assemblies were measured at more than 3 positions.
- **December 2014:**
  - 25 assemblies were measured (12 from the March campaign and 13 new ones).
  - All 4 corners were measured at position 138 cm.
- Two distinct types of assemblies: 10x10 and 8x8
- All BWR assemblies had cooling time ~> 7 years.

# Model functions to solve the inverse problem

---

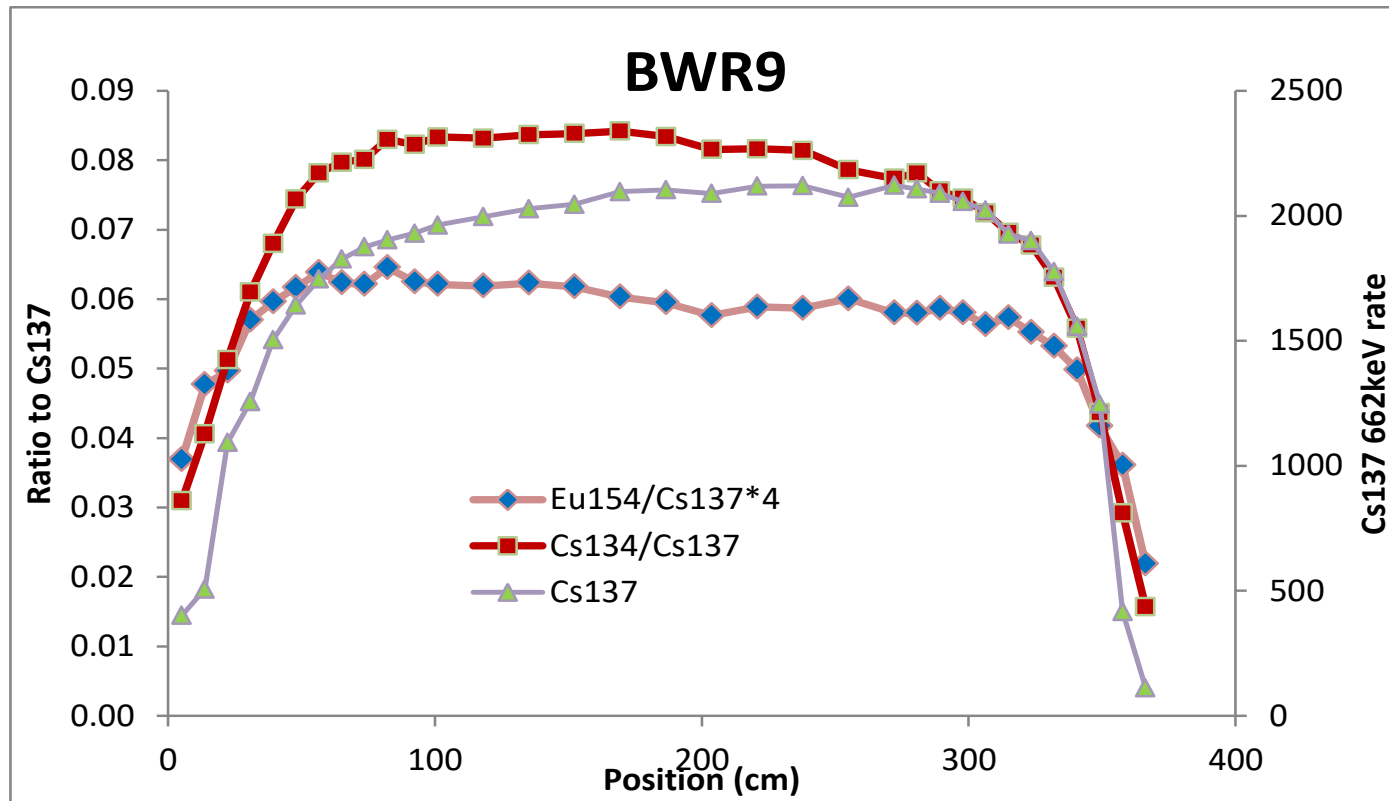
$$^{137}\text{Cs} = (aBU^b)e^{-\lambda_{137}CT}$$

$$\frac{^{154}\text{Eu}}{^{137}\text{Cs}} = (cBU^2 + dBU + e)e^{-(\lambda_{154} - \lambda_{137})CT}$$

$$\frac{^{134}\text{Cs}}{^{137}\text{Cs}} = (fBU^2 + gBU + h)e^{-(\lambda_{134} - \lambda_{137})CT} .$$

- The quadratic like function of the  $^{154}\text{Eu}/^{137}\text{Cs}$  and  $^{134}\text{Cs}/^{137}\text{Cs}$  ratios complicates the analysis
- Up to two possible ***BU*** solutions for each ratio

# Distribution as function of positions



BWR9 were measured at many positions along the assembly length

# BWR9 burnup results

Position (cm)	Burnup			Position (cm)	Burnup		
	Dec.	Mea.	Dev.		Dec.	Mea.	Dev.
5	7.95	8.65	8.8%	204	46.87	47.23	0.8%
14	17.17	11.88	-30.8%	221	47.35	48.07	1.5%
22	26.29	23.79	-9.5%	238	47.51	48.04	1.1%
31	30.53	27.62	-9.5%	255	47.35	46.78	-1.2%
39	34.37	33.77	-1.7%	272	47.65	48.00	0.7%
48	37.26	36.66	-1.6%	281	47.54	47.66	0.2%
56	39.40	39.12	-0.7%	289	47.35	47.13	-0.5%
65	40.98	41.14	0.4%	298	46.91	46.52	-0.8%
74	42.07	42.25	0.4%	306	46.08	45.65	-0.9%
82	43.02	42.95	-0.2%	315	44.90	43.40	-3.4%
92	43.75	43.73	0.0%	323	42.88	42.76	-0.3%
101	44.16	44.34	0.4%	332	40.38	39.92	-1.1%
118	44.74	45.10	0.8%	341	35.19	34.77	-1.2%
135	44.96	45.86	2.0%	349	26.75	27.56	3.0%
152	45.56	46.32	1.7%	358	12.99	8.59	-33.9%
169	46.25	47.50	2.7%	366	2.17	2.11	-2.7%
187	46.68	47.59	2.0%				

Dec. CT = 6.49y, Mea. CT = 6.30y, Dev. = -2.9%

# Previous work: PWR assemblies analysis



Contents lists available at [ScienceDirect](#)

Nuclear Instruments and Methods in  
Physics Research A

journal homepage: [www.elsevier.com/locate/nima](http://www.elsevier.com/locate/nima)



Determining initial enrichment, burnup, and cooling time of  
pressurized-water-reactor spent fuel assemblies by analyzing  
passive gamma spectra measured at the Clab interim-fuel  
storage facility in Sweden



A. Favalli <sup>a,\*</sup>, D. Vo <sup>a</sup>, B. Grogan <sup>e</sup>, P. Jansson <sup>c</sup>, H. Liljenfeldt <sup>e</sup>, V. Mozin <sup>f</sup>, P. Schwalbach <sup>d</sup>,  
A. Sjöland <sup>b</sup>, S.I. Tobin <sup>a</sup>, H. Trellue <sup>a</sup>, S. Vaccaro <sup>d</sup>

Nuclear Instruments and Methods in Physics Research A 830 (2016) 325–337



Contents lists available at [ScienceDirect](#)

Nuclear Instruments and Methods in  
Physics Research A

journal homepage: [www.elsevier.com/locate/nima](http://www.elsevier.com/locate/nima)



Passive gamma analysis of the boiling-water-reactor assemblies

D. Vo <sup>a,\*</sup>, A. Favalli <sup>a</sup>, B. Grogan <sup>e</sup>, P. Jansson <sup>c</sup>, H. Liljenfeldt <sup>e</sup>, V. Mozin <sup>f</sup>, P. Schwalbach <sup>d</sup>,  
A. Sjöland <sup>b</sup>, S. Tobin <sup>a</sup>, H. Trellue <sup>a</sup>, S. Vaccaro <sup>d</sup>



PWR paper  
published in  
Feb 2016 in  
NIM-A

BWR paper  
published in  
June 2016 in  
NIM-A



# A blind test

---

- We have one spectrum of a PWR SF pellet in hot cell from collaboration with Kaeri ~9 years ago.
- We analyzed the spectrum using the  $^{154}\text{Eu}/^{137}\text{Cs}$ ,  $^{134}\text{Cs}/^{137}\text{Cs}$ ,  $^{106}\text{Ru}/^{137}\text{Cs}$ , and  $^{144}\text{Ce}/^{137}\text{Cs}$  ratios.
- We sent the NIM-A article on PWR PG and the results of the analysis of the pellet to Kaeri and asked them to compare with the declared values.
- Analysis: BU = 65 GWd/tU, CT = 4.2 y, IE = 4.5%  $^{235}\text{U}$

# A blind test

---

- We have one spectrum of a PWR SF pellet in hot cell from collaboration with Kaeri ~9 years ago.
- We analyzed the spectrum using the  $^{154}\text{Eu}/^{137}\text{Cs}$ ,  $^{134}\text{Cs}/^{137}\text{Cs}$ ,  $^{106}\text{Ru}/^{137}\text{Cs}$ , and  $^{144}\text{Ce}/^{137}\text{Cs}$  ratios.
- We sent the NIM-A article on PWR PG and the results of the analysis of the pellet to Kaeri and asked them to compare with the declared values.
- Analysis: BU = 65 GWd/tU, CT = 4.2 y, IE = 4.5%  $^{235}\text{U}$
- Declared: BU = 65.2 GWd/tU, CT = 4.2 y, IE = 4.5%  $^{235}\text{U}$

# Conclusions and Remarks

---

- Passive gamma analysis can be used to determine BU, CT, and IE of spent fuel assembly. Assemblies with small CT give best results.
- Without any operator declaration, a 10 minutes measurement time of a PWR assembly can give the results for BU, CT, and IE with about 3% accuracy.
- BWR analysis is more complicated and less effective than PWR and the results are somewhat less accurate.
- A set of 4 measurements at 4 corners of an assembly can average out the variations of the assembly positions in the reactor during the burn cycles and can result in a better accuracy.